

REMARKS

Claims 100-105 and 107-120 are pending and, due to a restriction requirement, claims 109-119 are withdrawn from consideration. Claims 100-105, 107, 108, and 120 are rejected under 35 U.S.C. § 102(b) as being anticipated by Kirschhock et al. (Angew. Chem. Int. Ed. 40(14):2637-2640 (2001); “Kirschhock”). Applicants address each basis for rejection as follows.

Claim amendments

New claim 121 has been added. Claim 121 finds support, for example, in original claim 1, and at page 12, lines 1-27, of the WO 2005/000740 publication. No new matter has been added by the present amendment.

Rejection under 35 U.S.C. § 102(b)

Claims 100-105, 107, 108, and 120 are rejected under 35 U.S.C. § 102(b) as being anticipated by Kirschhock. In particular, the Office states (pages 2 and 3):

In response [to applicants’ arguments], the prior art assembly of nanoslabs is described as being a pin-in-hole type assembly. Tetrapropyl ammonium (TPA) has four propyl groups that provide the “pins” and the pore openings on the face of the nanoslabs are the “holes.” This is illustrated in Figure 6.

* * *

The assembly of the slabs in the prior art is thus likened to assembling wood blocks with dowels. The TPA is similar to a small sphere (the nitrogen) with attached dowels (the propyl groups). The nanoslabs are like wood blocks with holes (pores) that receive the dowels. Following this analogy, the wood blocks cannot be assembled flush, because the knob in the middle of the dowels keeps them apart. In the same manner, the nanoslabs cannot be assembled flush, because the nitrogen atom requires space between the slabs. This space is the second level of porosity.

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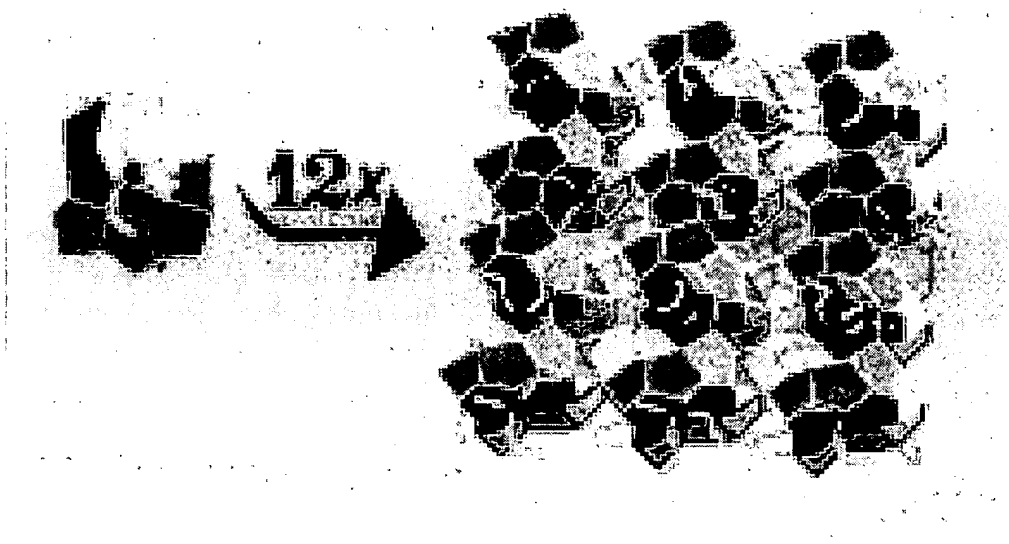
In an alternative interpretation, the zeolites themselves have a single layer of porosity. When assembled, however, the pores on the surface of the nanoslabs are partially obstructed by the propyl “pins” on TPA. These surface pores therefore have a different amount of volume from other pores, creating a second level of porosity.

The independent claims, claims 100 and 107, read as follows.

100. A crystalline mesoporous silica material being obtained by assembly of nanometer size building units having zeolite framework, said crystalline mesoporous silica material having two or more levels of porosity and structural order, and wherein the internal structure of said nanometer size building units does not give rise to Bragg type diffraction in a powder X-ray diffraction pattern of said crystalline mesoporous silica material.

107. A process for making a crystalline mesoporous silica material, said crystalline mesoporous silica material being obtained by assembly of nanometer size building units having zeolite framework, said crystalline mesoporous silica material having two or more levels of porosity and structural order, wherein said building units are nanoslabs of substantially uniform size having the Silicalite-1 zeolite framework, comprising the steps of (a) generating said nanoslabs having said zeolite framework by tetrapropylammonium-ion mediation and (b) assembling said nanoslabs through interaction with a cationic surfactant or triblock copolymer molecule

Applicants respectfully submit that the Office appears to be misinterpreting the assembly of nanoslabs disclosed in Kirschhock, because the nitrogen and four propyl groups are all occluded within the pores and the nitrogen does not obstruct the assembly process, as stated by the Office. Tetrapropylammonium (TPA)-containing precursors are the building blocks in the crystallization of silica. In the first steps, slab-shaped silicalite nanoparticles are formed by ordered combination of the precursors (see the below figure from Kirschhock provided in the hyperlink included in the present Office Action).



The nanoslabs have MFI-type zeolite framework topology and play a key role in TPA-ion-mediated zeolite crystallization from monomeric and polymeric silica sources.

The clicking together of nanoslabs leads to the connection of the channel segments from the two units and formation of a larger channel out of the two short segments present in the original units. When these nanoslabs are connected to one another, they become indistinguishable from one another and form a continuous silicate network in which it is impossible to find the original plane of fusion i.e., they fit completely flush. In this continuum there is only one type of pore system. The nitrogen and the propyl arms of the TPA are all in the pores, i.e., do not cause the obstruction stated by the Office, and hence cannot give rise to the second level of porosity suggested by the Office.

The porosity is caused by the formation of a network of bonded silicon and oxygen atoms with wide rings that have free openings that typically measure 1 nm or less. The rings line up

and make channels and cages. Nanoslabs contain such very short channels. Figure 6 of Kirschhock (see the figure inserted above) shows the linking of nanoslabs. In all the linkage variants shown in Figure 6 (linking through corners, edges and faces) the framework of one slab is continued in the slab connected to it. By doing so, the channels get longer, but no new type of pore is created. The channel diameter remains the same. The porosity of the zeolite is that determined by the specific zeolite framework type. In this instance, it is the “MFI” type. A zeolite represents the most compact manner of stacking nanoslabs. Kirschhock thus teaches how to make a compact stacking of nanoslabs. This compact stacking and coalescence leads to the formation of a zeolite crystal, which displays X-ray diffraction at small lattice spacings (d-values smaller than 1.5 nm). Therefore, an inherent property of zeolites, including those disclosed in Kirschhock, is that they exhibit only one level of porosity and the inherent presence of two or more levels of porosity, as suggested by the Office, can be excluded.

The present specification teaches how to make materials with more open structures than those disclosed in Kirschhock, in which free spaces - several nanometers wide - are left between individual nanoslabs. In these new materials, the smallest pores are identical to those present inside the nanoslabs and inside the zeolite disclosed in Kirschhock. This is the first-level of porosity. By assembling the nanoslabs systematically and leaving void spaces between them, a well-defined secondary porosity is created. Stacking patterns of higher order can even be imposed on a third level, and so on. Kirschhock does not teach how to assemble nanoslabs such that systematically open spaces are left in between.

In support of the differences between the presently claimed crystalline mesoporous silica

material and the material disclosed by Kirschhock, Applicants direct the Office's attention to the enclosed Declaration by Dr. Alexander Aerts, an inventor of the presently claimed invention.

Here, in comparing a crystalline mesoporous silica material encompassed by the present claims (Zeotile-4) with a material made according to the methods disclosed in Kirschhock (Silicalite-1),

Dr. Aerts states (paragraph 3; EXPERIMENT C):

The Silicalite-1 crystals only exhibited microporosity as indicated by the upturn in the relative pressure range corresponding to micropore filling but no upturn in the relative pressure range corresponding to mesopore filling. [See Figure 1 in the Declaration.] Silicalite-1 crystals thus exhibit a single level of porosity i.e., microporosity.

On the other hand, Zeotile-4 exhibited microporosity as indicated by the upturn in the relative pressure range corresponding to micropore filling and mesoporosity as indicated by the upturn in the relative pressure range corresponding to mesopore filling as indicated by a sharp upturn around $p/p^{\circ} = 0.75$. This upturn is due to capillary condensation of nitrogen in mesopores with a narrow pore size distribution. The adsorption and desorption isotherms show hysteresis typical for cylindrical shaped mesopores. Barrett-Joyner-Halenda (BJH) analysis yielded an average mesopore diameter for Zeotile-4 of 8.3 ± 0.3 nm (Figure 1, right). Alpha-s analysis revealed that the mesopore volume of Zeotile-4 was 1.33 mL/g. Zeotile-4 material thus exhibit two levels of porosity: microporosity and mesoporosity.

Although Zeotile-4 crystals and Silicalite-1 zeolite crystals are both prepared starting from nanoslab sol, Silicalite-1 only exhibited microporosity due to the nanoslabs being densely packed, whereas the Zeotile-4 exhibited both microporosity and mesoporosity.

The results shown in EXPERIMENT C and Figure 1¹ of the Declaration show that the zeolite prepared according to the process of Kirschhock (Silicalite-1) is microporous, i.e., exhibits a single level of porosity, whereas the crystalline mesoporous silica material encompassed by the present claims (referred to as Zeotile-4 in the Declaration), is microporous and mesoporous, i.e.,

¹ A copy of Figure 1 in color is enclosed as a courtesy to the Office.

exhibits two levels of porosity. The claims require the crystalline mesoporous silica material to have two or more levels of porosity and structural order. Clearly, as evidenced by the Declaration, the material disclosed in Kirschhock does not meet all of the limitations of the present claims and, therefore, claims 100-105, 107, 108, and 120 cannot be anticipated by Kirschhock.

Applicants further note that the new materials disclosed in the present application exhibits X-ray diffraction peaks at larger d-values than the zeolite disclosed in Kirschhock owing to the systematic stacking of nanoslabs in an open manner rather than the coalescence of the nanoslabs into a continuous crystalline lattice characterized by small d-spacings (see Figure 1A of the specification and Figure 5 of Kirschhock).

Moreover, the presence of sodium ions, and TPA in the synthesis mixture for preparing nanoslabs are sufficient for synthesizing a zeolite as disclosed in Kirschhock, but not for synthesis of the claimed new materials having more than one order of porosity. For creating the secondary porosity, surfactant or polymer molecules are needed. These molecules occupy the spaces between the nanoslabs thereby interfering with the assembly of the nanoslabs to form a continuum. The surfactants and polymers are organic molecules are then removed by calcination giving rise to the second level of porosity. Tetrapropylammonium and tetrabutylammonium ions used in the synthesis of the nanoslabs themselves, cannot assume the role of template for the creation of the secondary porosity, because the nitrogen and the propyl arms of the TPA are all in the pores of the first level of porosity. The surfactants for assembling the nanoslabs typically have a long alkyl chain, such as in cetyltrimethylammonium ions.

For all the above reasons, Applicants submit that the crystalline mesoporous silica material encompassed by claims 100 to 105, 107, 108 and 120, as well as by new claim 121, is free of the anticipation rejection under 35 U.S.C. §102 (b) over Kirschhock. This basis for rejection should be withdrawn.

CONCLUSION

Applicants submit that the claims are now in condition for allowance, and such action is respectfully requested.

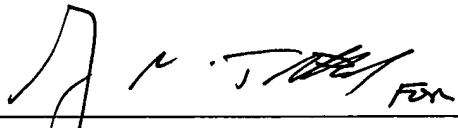
If there are any charges or any credits, please apply them to Deposit Account No. 03-2095.

Respectfully submitted,

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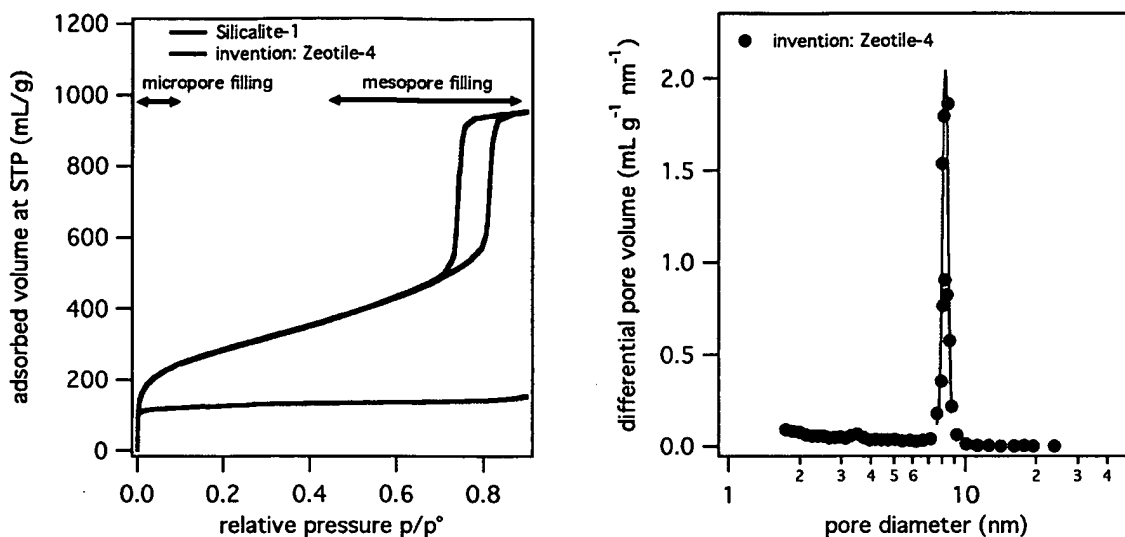


Figure 1. (left) Nitrogen physisorption isotherms of Zeotile-4 and Silicalite-1 crystals. The pressure ranges in which micropores and mesopores are filled are indicated. (right) Mesopore diameter distribution of Zeotile-4 calculated from the desorption isotherm using the BJH model.